Hard X-ray Science with a Diffraction-Limited Storage Ring

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Outline

- Technique opportunities arising from transverse coherence
- Science:
  - Energy storage materials
  - Materials processing and mesoscale assembly
  - Fluctuations and dynamics in biological materials
  - Integrating new functionalities in electronics
- Conclusion

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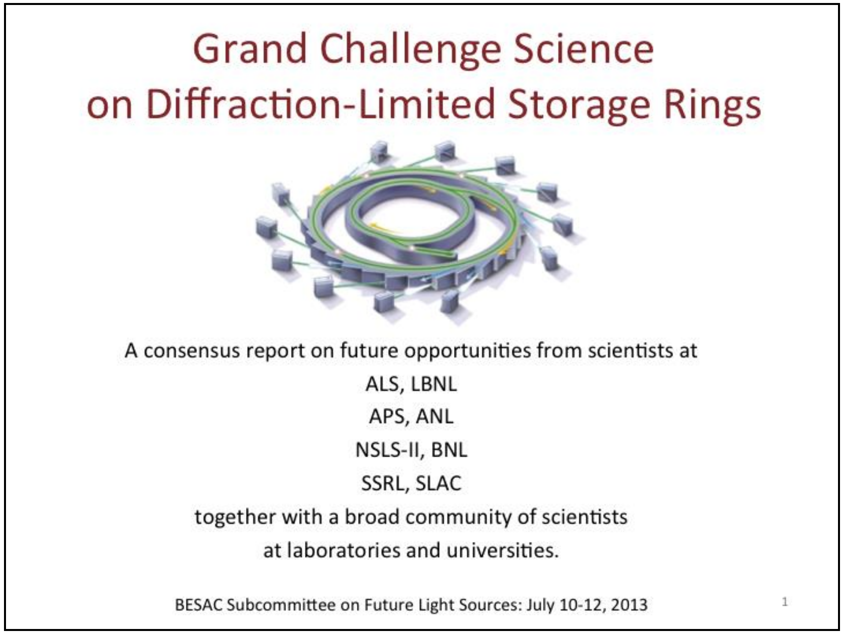
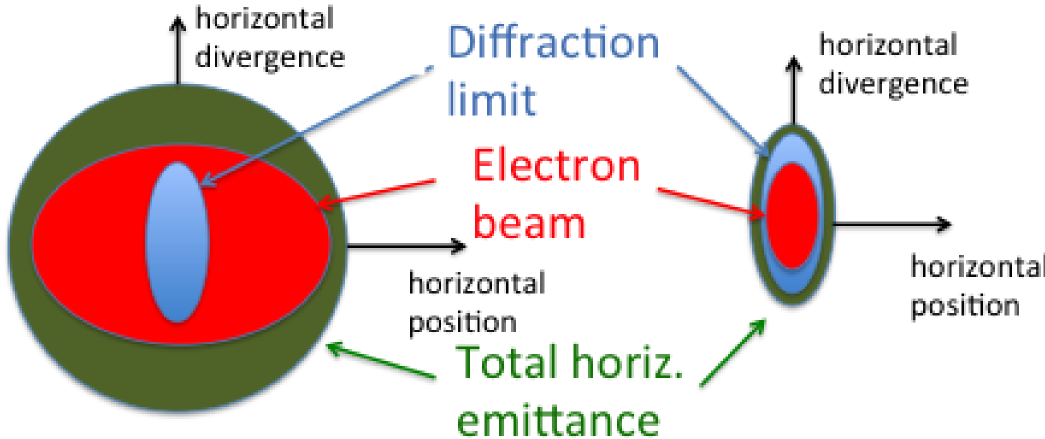


Stephan Hruszkewycz, Paul Fuoss, Paul Fenter, Dean Haeffner, Martin Holt, John Freeland, Hoydoo Yoo, Oleg Shpyrko, Stefan Vogt, Juan de Pablo, Ian McNulty, Michael Pierce…

UW-Madison: Qingteng Zhang, Joonkyu Park, Pice Chen

Transverse Coherence of Diffraction Limited Storage Ring Sources

- **Fundamental limit of source size and divergence depends only on wavelength**



$$\varepsilon_r = \text{diffraction limited emittance} = \sigma_\gamma \sigma'_\gamma = \frac{\lambda}{4\pi} = \begin{cases} \text{80 pm @ 1 keV} \\ \text{8 pm @ 10 keV} \end{cases}$$

- **Coherent fraction = ratio of diffraction-limited emittance to total emittance**

$$f_{coh} = \frac{F_{\mathrm{coh,T}}(\lambda)}{F(\lambda)} = \frac{\sigma_\gamma \sigma'_\gamma}{\underbrace{\sigma_{\mathrm{Tx}} \sigma_{\mathrm{Tx}'}}} \frac{\sigma_\gamma \sigma'_\gamma}{\sigma_{\mathrm{Ty}} \sigma_{\mathrm{Ty}'}}$$

Hard x-rays: atomic-scale wavelength/high momentum, K and L edge atomic edges, weak interaction with matter.

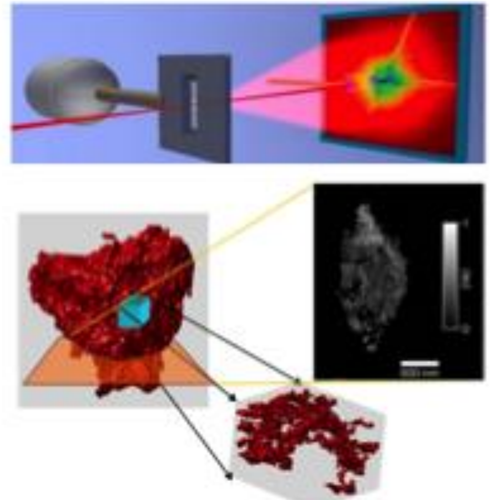
Three Ways to Use Transverse Coherence

- Intense Focused Beams for Local Probes
- Microscopy via Scanned Probes/Ptychography/Coherent Diffraction Imaging
- Fluctuations and Dynamics

Coherence of DLSR's Provides a Vast Improvement in X-ray Imaging

Coherent Diffraction Imaging

- Resolution limited by wavelength and sample stability – not optics.
- Recover real and imaginary parts of refractive index: magnetization, composition, bonding configuration.
- Challenge is to reach atomic scale.

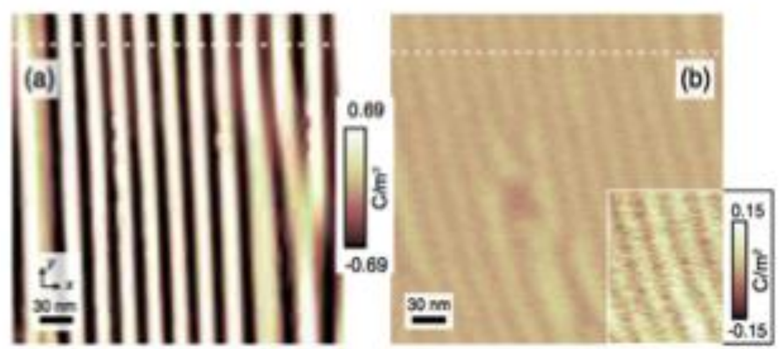


Nanofoam Diffraction Pattern/Reconstruction

A. Barty, *et al.*, Phys. Rev. Lett. **101**, 055501 (2008)

Wavelength-Resolution Ptychography

- Scanned-beam **ptychography** adapts CDI to continuous samples, resolution far better than focused x-ray spot size.
- Improved coherence of DLSRs will allow coherent imaging techniques to reach wavelength resolution.

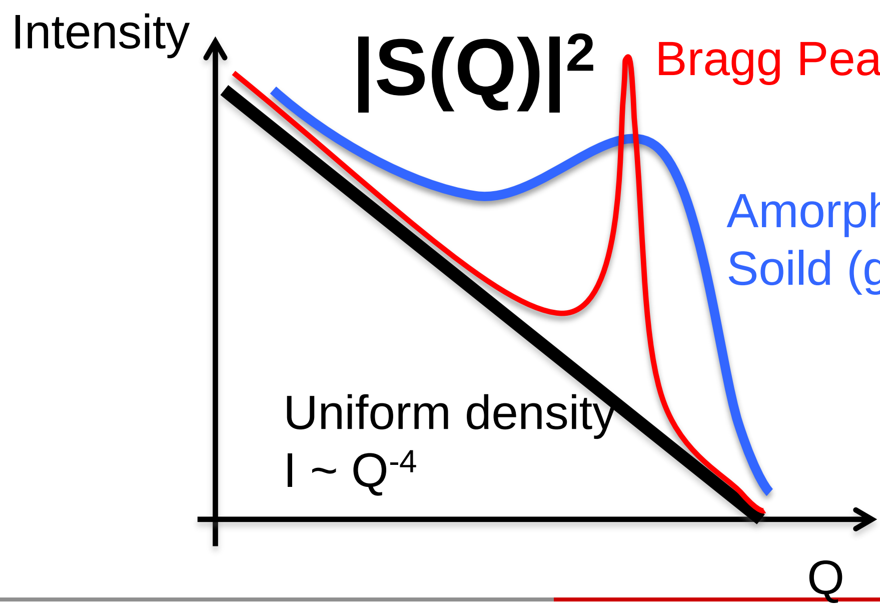


Polarization Domains in $PbTiO_3$

Hruszkewycz *et al.*, PRL (2013)

Hard X-ray Imaging

- Image using spot size that matches problem.
- Scan or reconstruct from S(Q).
- Resolution, information, depends on S(Q)
- Density, composition, magnetism, strain…

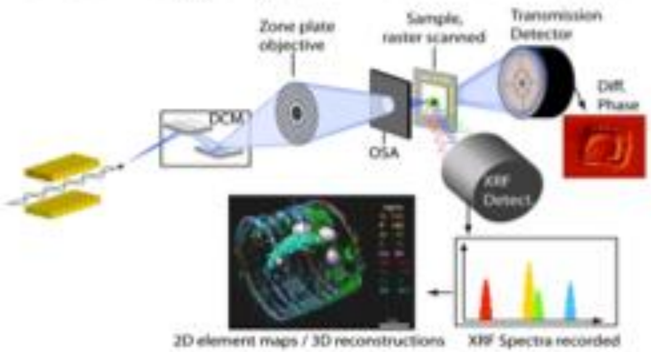


Oleg Shpyrko

A New Regime of Scattering and Spectroscopy with Nanobeams: *nanoXAS, nanoXRF, nanoARPES/APXPS, nanoRIXS*

X-ray fluorescence nano-tomography

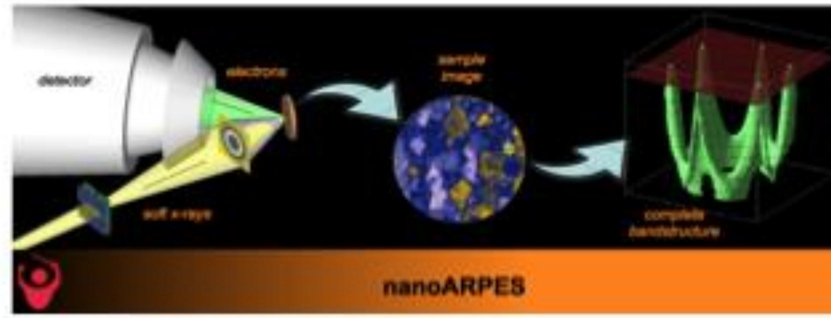
3D elemental mapping of functional mesostructures



de Jonge, et. al., PNAS 107, 15676 (2010).

nanoARPES

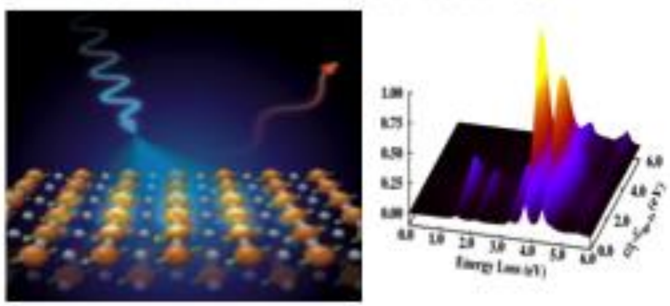
Electronic texture and single particle response



Eli Rotenberg, ALS

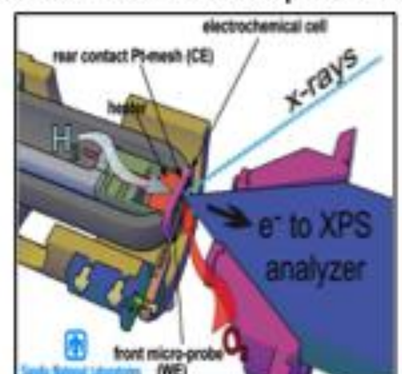
nanoRIXS

Understanding coupled excitations in heterogeneous materials and nanostructures



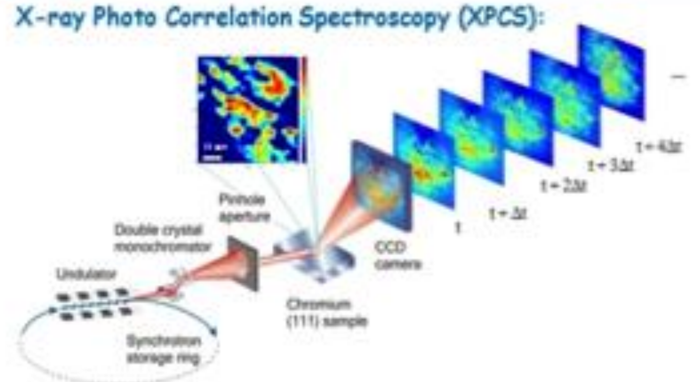
nano-APXPS

in operando studies of complex interphases



DLSRs will vastly expand the capability and capacity of **scanned** x-ray probes: high flux at resolution approaching 1 nm.

Fast Fluctuations with XPCS



X-ray photon correlation spectroscopy

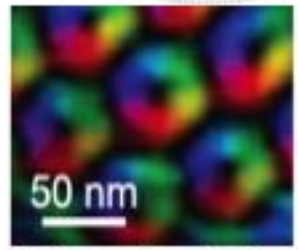
- Chemical, magnetic, and structural fluctuations

Accessible time scale proportional to (coherent flux)^2

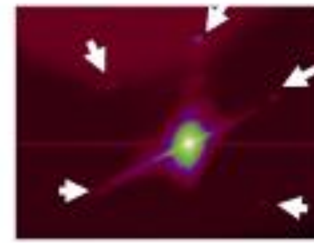
- 100-fold increased brightness improves time resolution by 10^4

DLSRs enable ns-resolution studies of nm-scale fluctuations

- Reaction-diffusion
- Self-assembly
- Domain wall motion
- Complex order parameters



Skyrmion lattice.
*Seki et. al. Science **336**, 198 (2012).*



Skyrmion at Cu L_3 edge
S. Roy, ALS, 05/2013

Time to probe 1 ns fluctuations:

Today:	50,000 hours
DLSR + modern BL:	0.05 hours

Dream: Correlation within a single pulse, 10 ps fluctuations:

DLSR + new BL + detector: ~2 hours!!

DLSRs will revolutionize our ability to probe **fluctuations** at molecular length scale and subnanosecond timescales.

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Opportunity

- Electronic, chemical and atomic structure of energy storage materials control their function.
- Structure evolves during operation and use new understanding to optimize durability, capacity, and discharge rate.

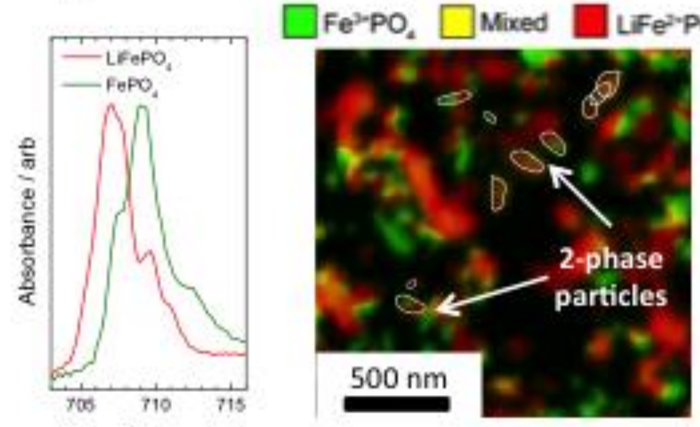
Challenge

- Requires *in operando* nm-resolution chemical and spectroscopic imaging and structure determination.

Unique DLSR Strengths

- *in operando* 3D tomographic chemical mapping using ptychography/CDI.
- Nanofocusing for *in-situ/in-operando* experiments and use of a **multi-modal** approach.

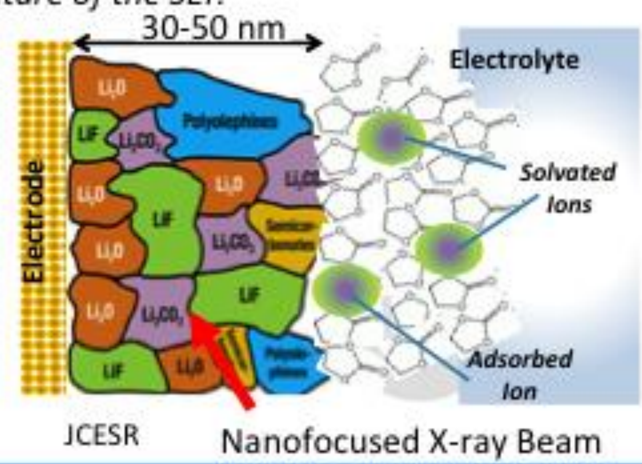
Resolving Oxidation States, Chemical Functionality



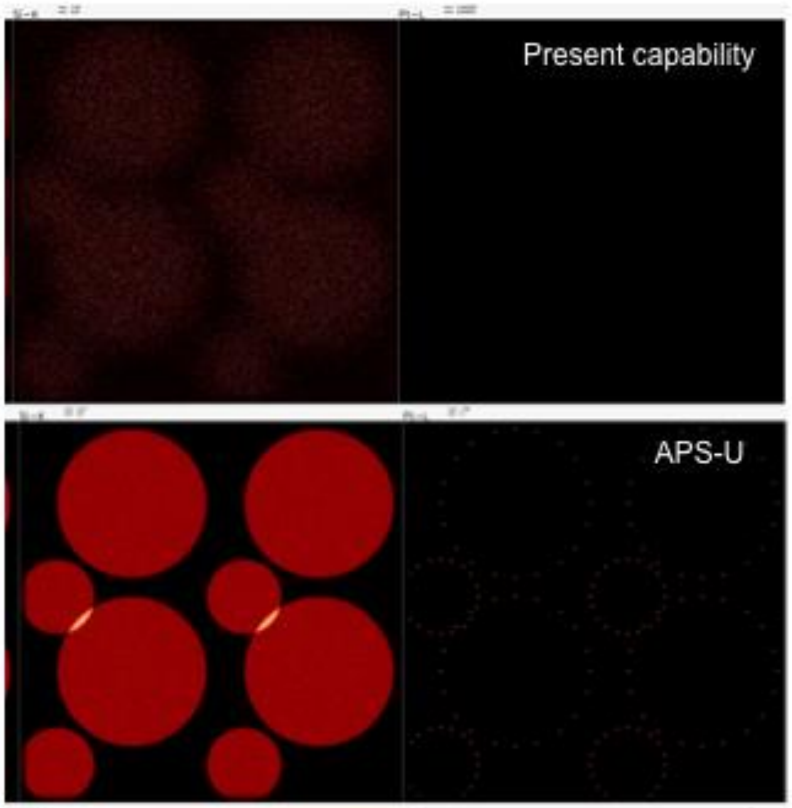
Chueh, et. al., *Nano. Lett.* **13**, 866 (2013)

Solid-Electrolyte Interface: Structure and Function

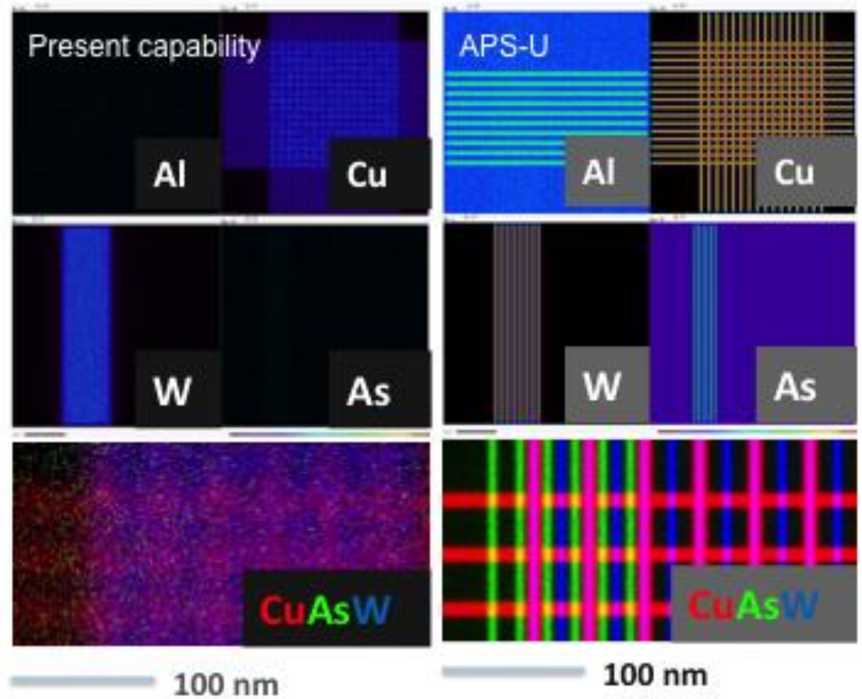
What controls the formation, composition, and structure of the SEI?



Fluorescence Nanoprobe



Simulated Si particles with 1 nm^3 Pt islands as model for catalysis. Left panels show Si x-ray fluorescence; right panels show Pt fluorescence



Simulated x-ray fluorescence scanning of semi-conductor materials; green = single atomic layer of As

Stefan Vogt

With DLSR it will be possible to resolve individual 1 nm Pt catalyst particles.

Requires: Small focal spot size, high focused flux.

Controlling Materials Exhibiting Nanoscale Order

Opportunity

- New electronic, mechanical, optical physics and devices require control of nanoscale order in electronic, magnetic, and orbital degrees of freedom.
- Non-equilibrium theory beginning to emerge via density functional theory but experiments have been challenging.

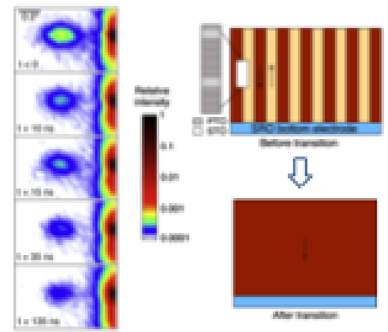
Challenge

Now use the macroscopic averages of nanoscale order parameters (e.g. ferroelectric or spin/orbital stripes). Local characterization and control not yet possible.

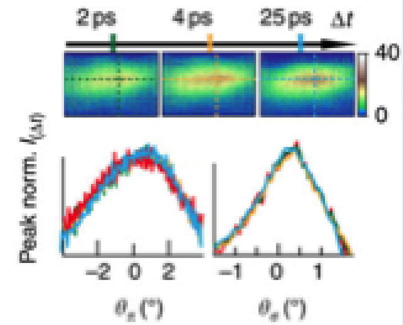
DLSR Strength

Higher coherent flux, allowing us to probe equilibrium dynamics and transitions driven by external fields.

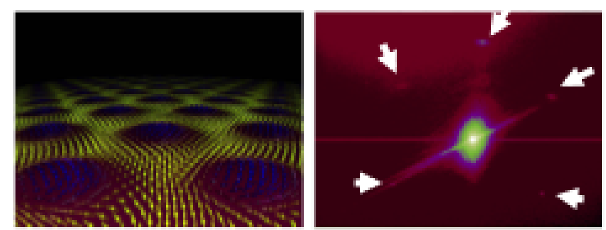
“Soft” Hard Materials: Nanoscale Order



Ferroelectric Stripe Nanodomains



Nickelate Stripes

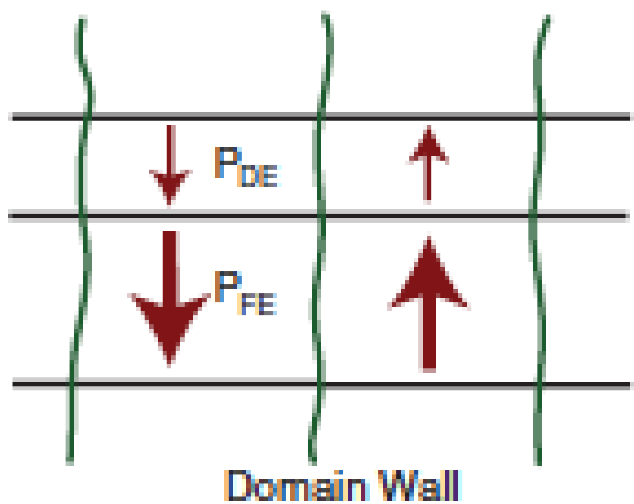


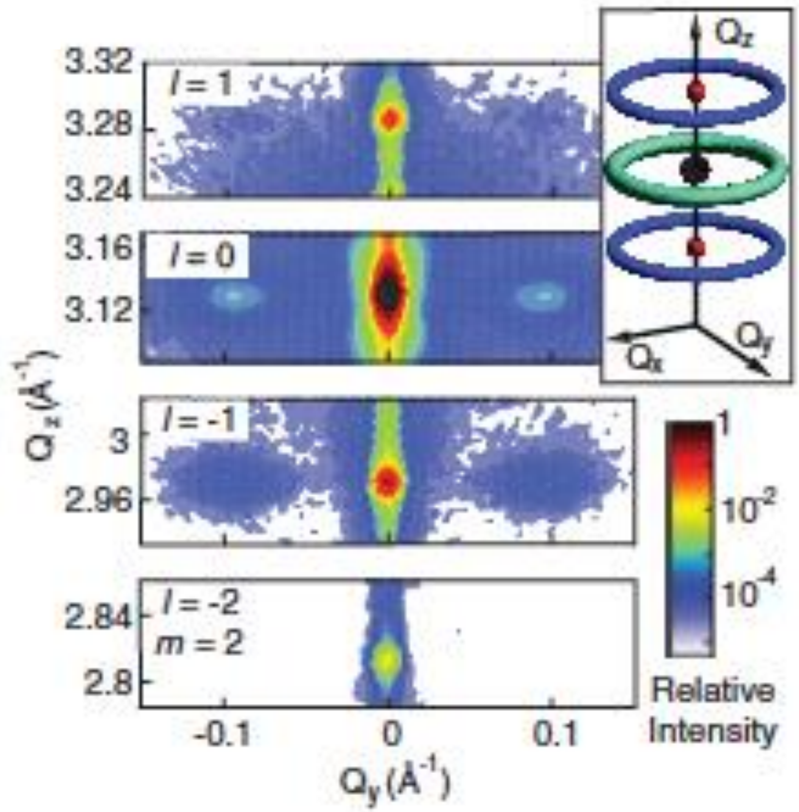
Skyrmion Lattice

Real-time coherent diffraction imaging, ptychography, XPCS.

Nanodomains in Ferroelectric/Dielectric Superlattices

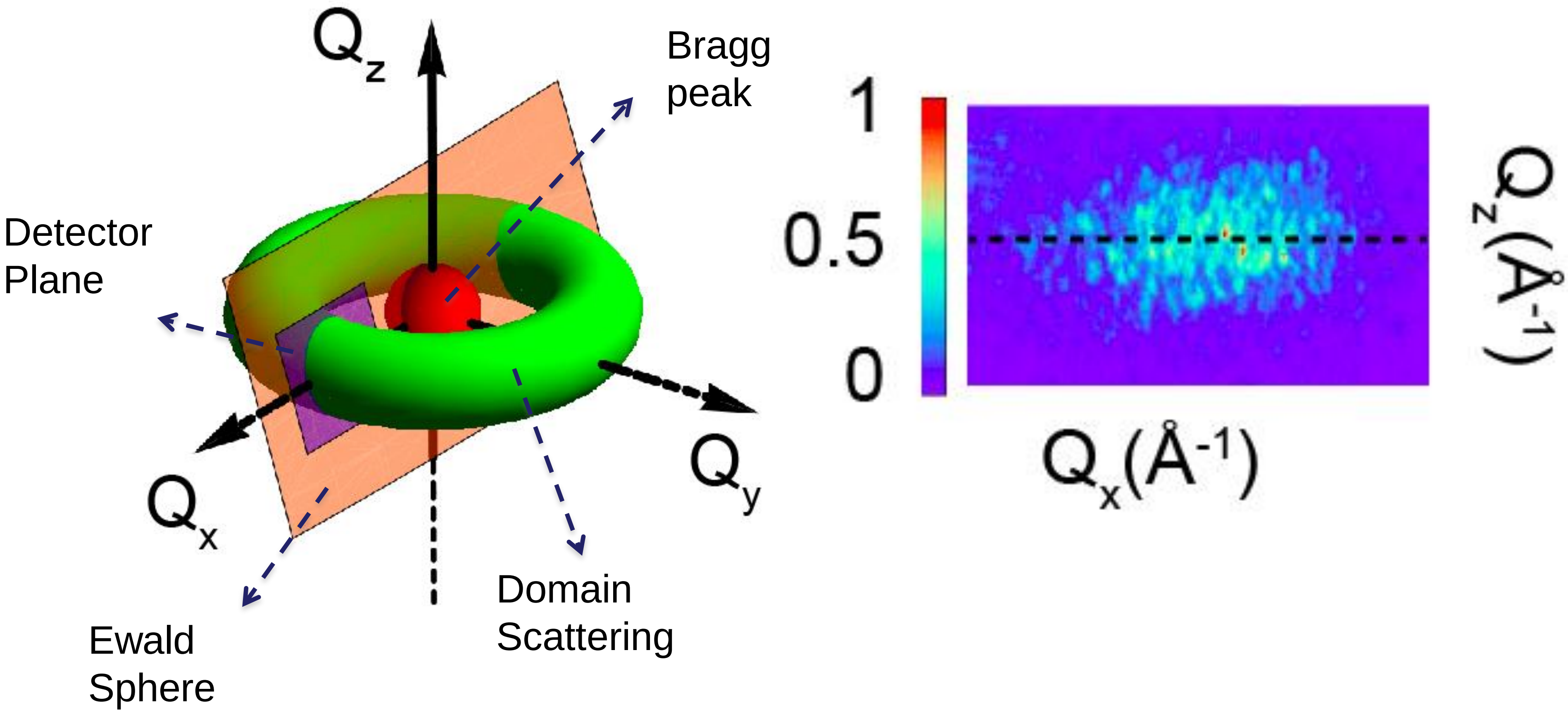
- Spontaneous Nanoscale Structural and Electronic "Order"



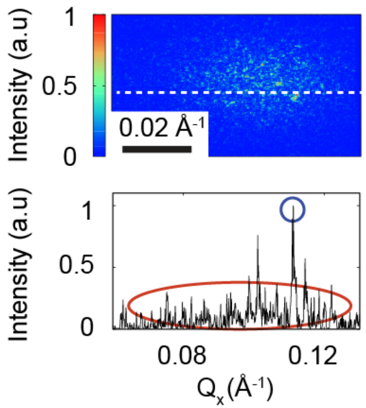


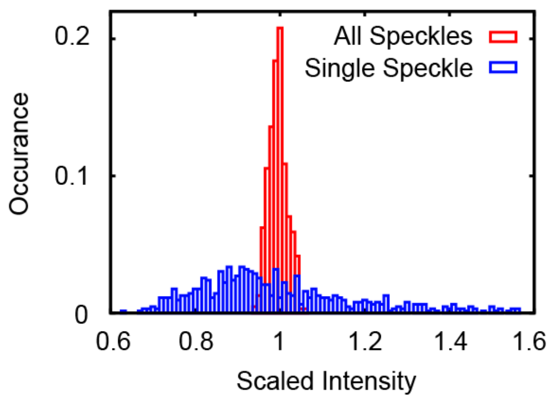
P. Chen, *et al*., Phys. Rev. Lett. **110**, 047601 (2013)

Domain Coherent Scattering



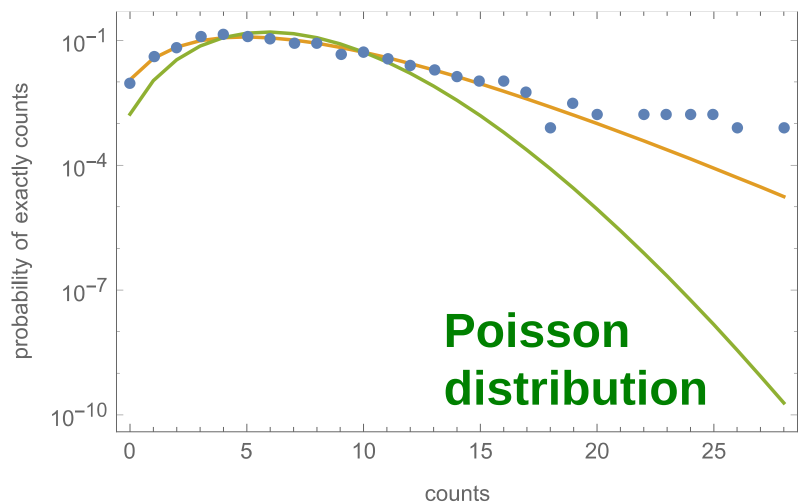
Speckle Statistics





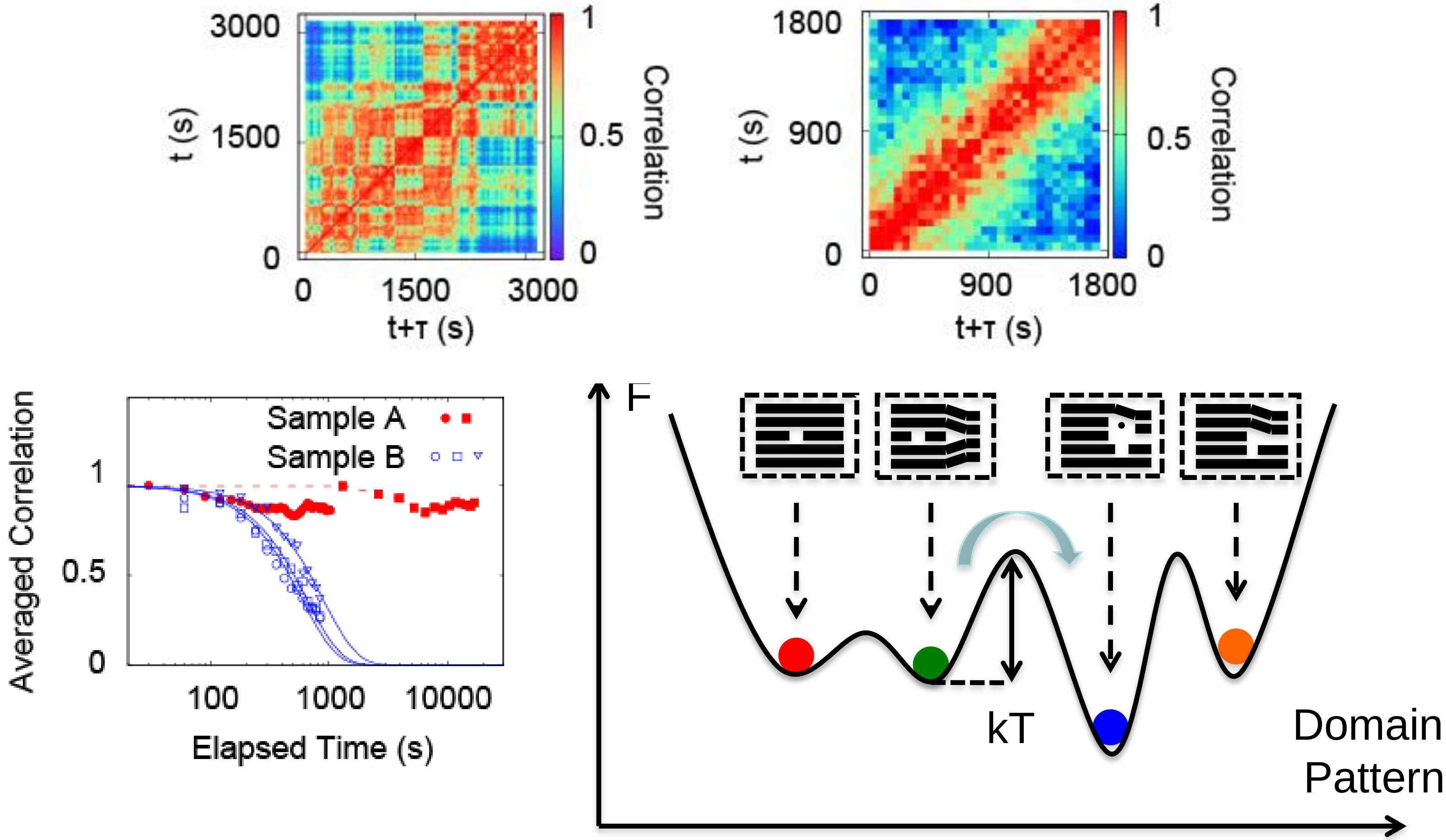
- Speckle Intensity Depends on Beam Location
- Average Domain Diffuse Scattering Constant

- Intensity statistics match speckle distribution



Binomial distribution
(e.g. Hruszkewycz, Phys. Rev. Lett. 109, 185502 (2012))

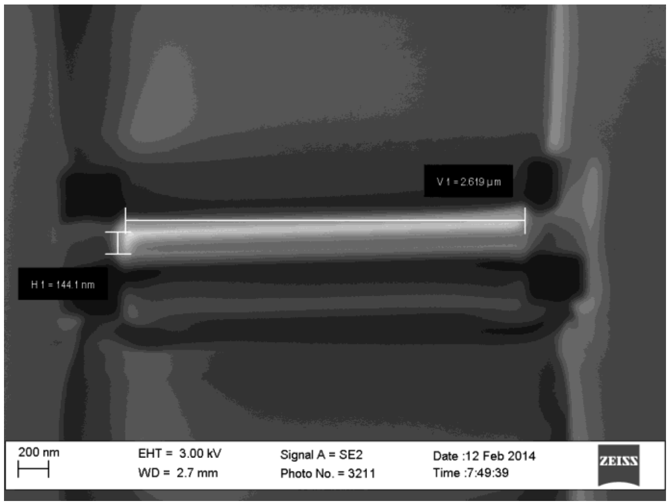
Decorrelation: "Soft" Domain Dynamics

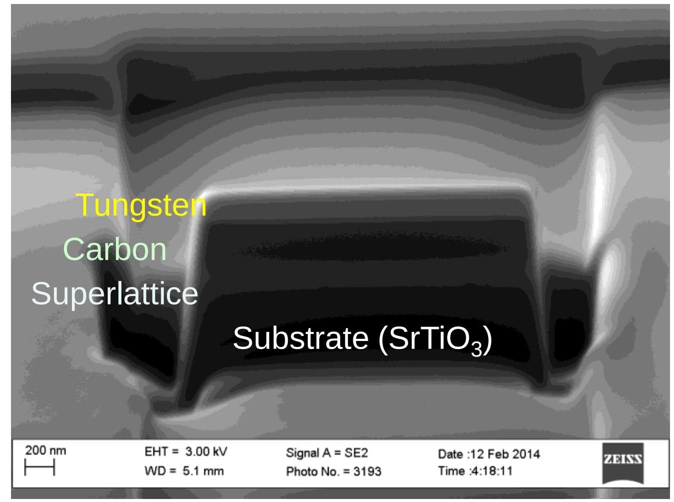


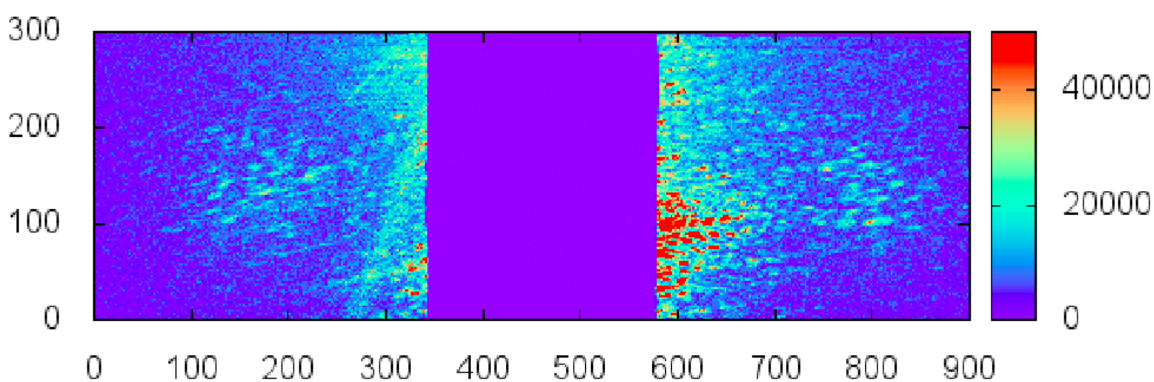
Slow long-lengthscale rearrangement dynamics.

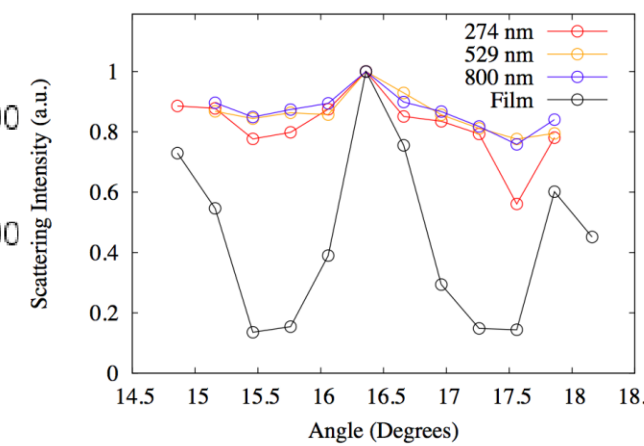
DLSR will allow fluctuations to be studied at much shorter timescales.

Domains in Restricted Geometries









144 nm island domain coherent scattering

Structure and Dynamics of Mesoscale Assembly

Opportunity

- Materials processing and assembly require dynamics far from equilibrium.
- Interfaces (e.g. liquid/air, polymer/polymer) induce assembly always in dynamic states.

Challenge

- Now have snippets of information, too slow to capture the crucial dynamics.
- Reaction kinetics during the self-assembly process on are nm- and ns-µs, 3-4 orders of magnitude faster dynamics in electrolyte than in vacuum, difficult to investigate at existing SR.

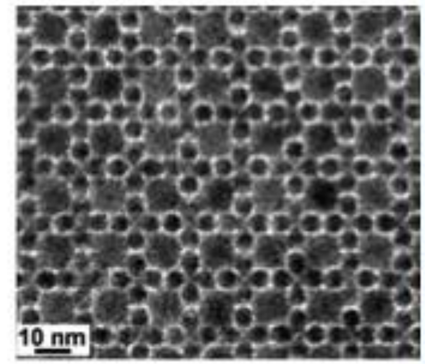
DLSR Strength:

- Coherent beam allows focal spot size to be freely chosen to match system.
- Interrogate the structure evolution at fundamental with coherent x-ray scattering or imaging.

Nanoparticle Assembly

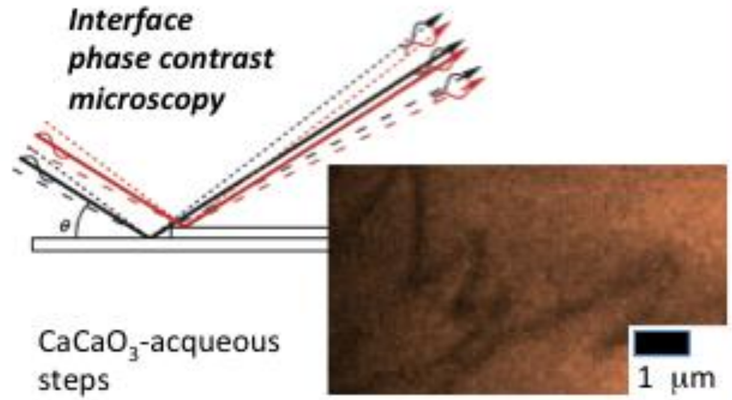
Mesoscale structures for high-density photonics, storage media and nanolithography.

Atomic to macroscopic



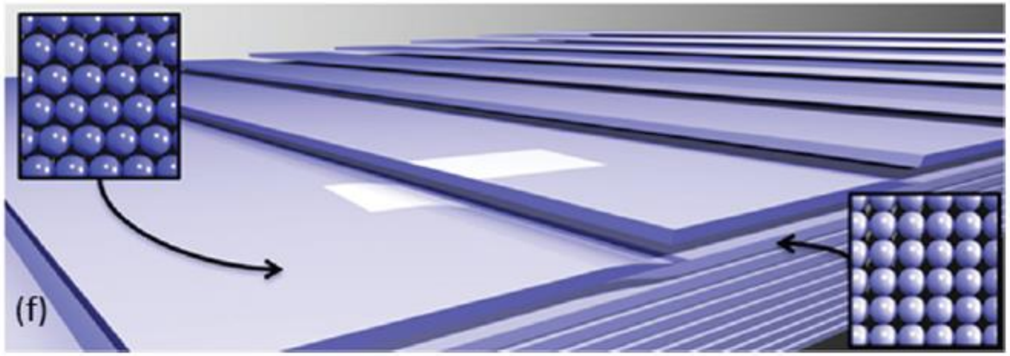
Coherent Scattering and Interface Microscopy in Continuous Processes

Interface phase contrast microscopy

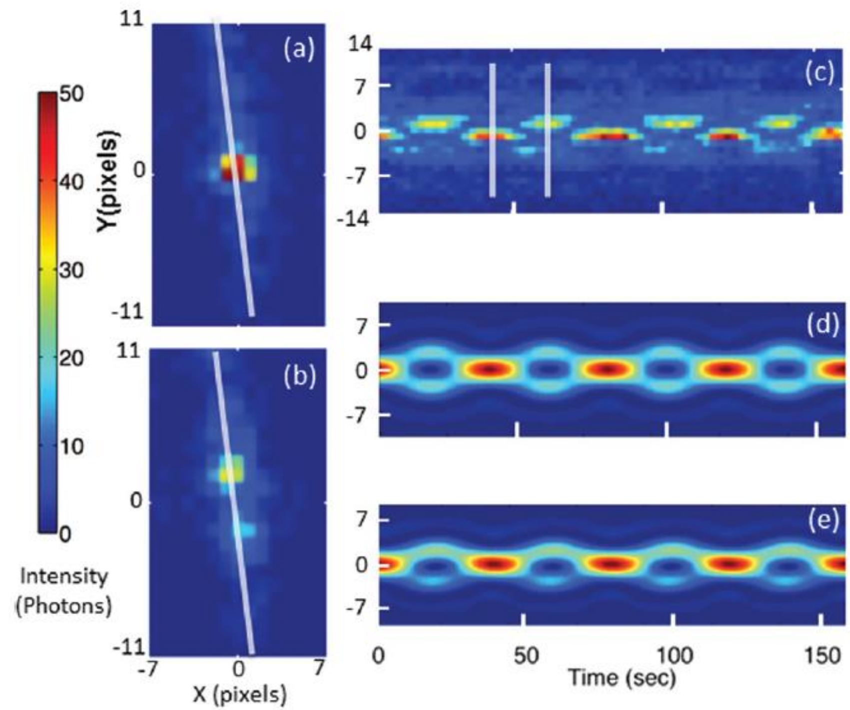


CaCaO$_3$-acqueous steps

Now: Step Flow in "Ideal" Systems



Pt (001) step flow during evaporation



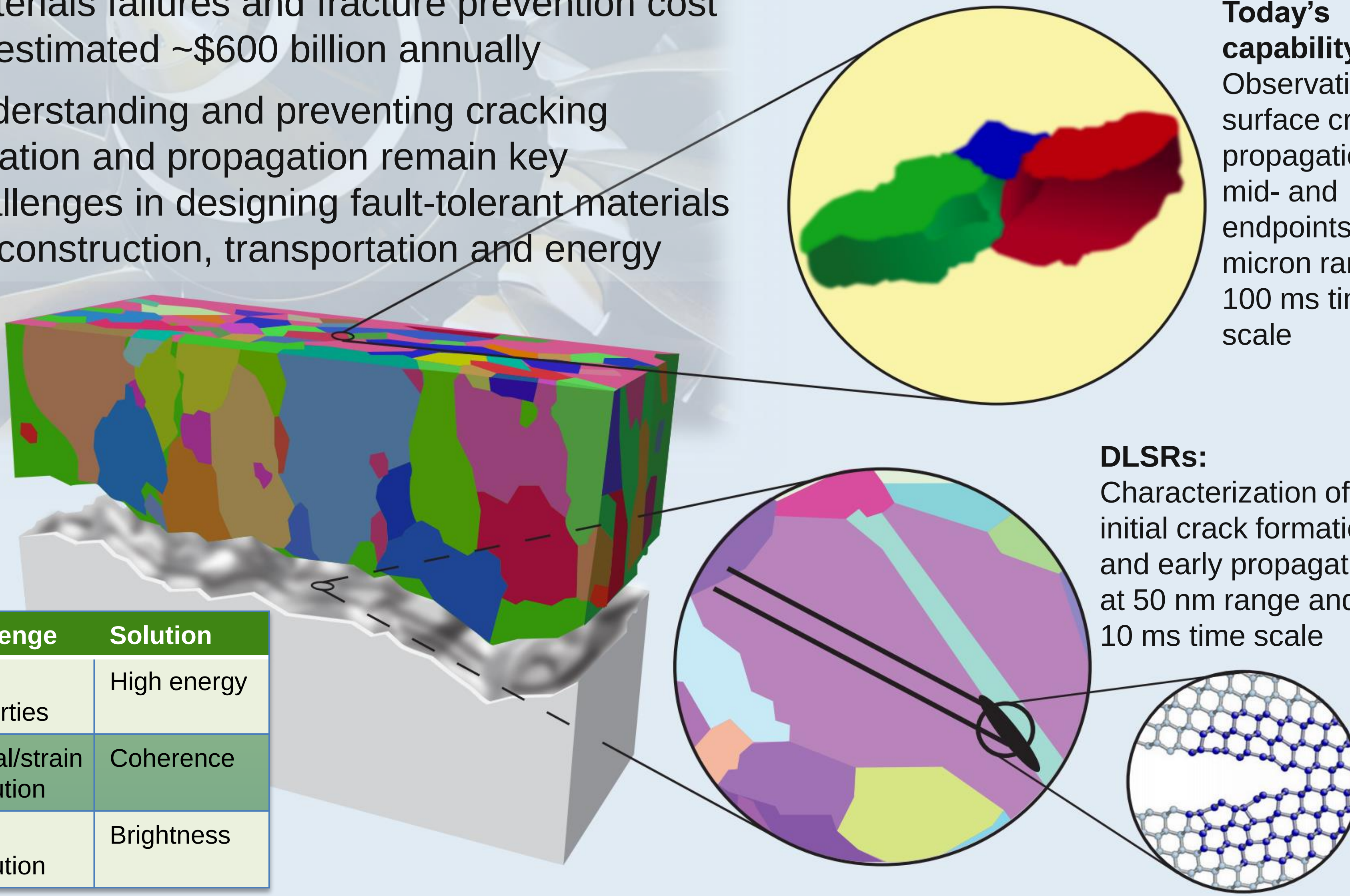
Oscillations in coherent scattering

M. S. Pierce, *et al*., Appl. Phys. Lett. **99**, 121910 (2011).

Materials science: Dynamic studies of crack initiation

Materials failures and fracture prevention cost an estimated ~$600 billion annually

Understanding and preventing cracking initiation and propagation remain key challenges in designing fault-tolerant materials for construction, transportation and energy



Today's capability: Observation of surface crack propagation at mid- and endpoints, at 1 micron range, 100 ms time scale

DLSRs: Characterization of initial crack formation and early propagation, at 50 nm range and 10 ms time scale

Challenge	Solution
Bulk properties	High energy
Spatial/strain resolution	Coherence
Time resolution	Brightness

Nanoscale Collective Dynamics in Biomaterials

Opportunity

- *Biological function requires motion*: ps-to-s scale non-thermal reorganization.
- *Structure-dynamics-function relationships* for *model artificial* and *biomembranes*.

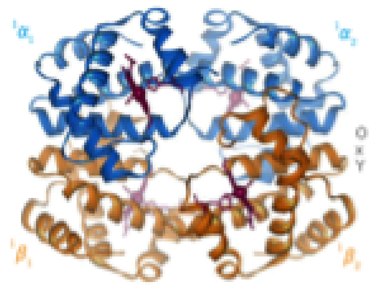
Challenge

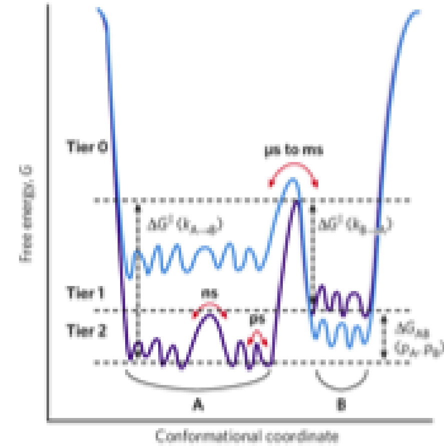
- Many degrees of freedom in protein backbones and side chains.
- Collective fluctuations occur on a wide range of time scales from ps to s.
- Functional systems are not periodic.

DLSR Strength

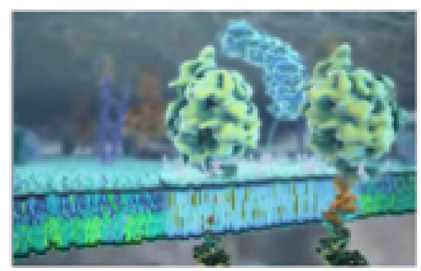
- Simultaneous probe of nanoscale order, assembly, and dynamics via microscopy and scattering.
- Multimodal approach (CDI, TXM, SAXS, XPCS, cryo-TEM) to enhance spatial & temporal resolution and chemical speciation

Collective Dynamics of Proteins

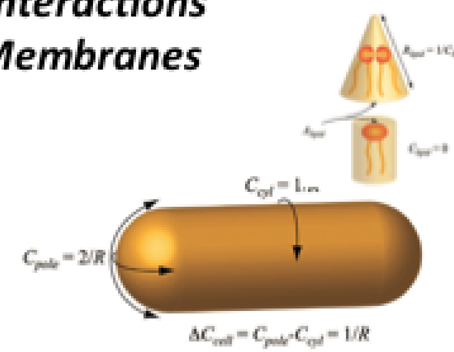




Elastically Mediated Interactions between Proteins in Membranes



Harvard MCB Inner Life of a Cell



$$\Delta E_{curv} \approx \kappa C_{lipid} \Delta C_{cell} A_{lipid}$$

Rare Events in Complex Non-harmonic Energy Landscapes

Opportunity

Non-equilibrium systems have dynamics with important rare events.

Fluids, glasses, biological dynamics, materials assembly and nanostructure dynamics, long-range order in emerging electronic materials.

Need to bridge kinetics and dynamics.

Challenge:

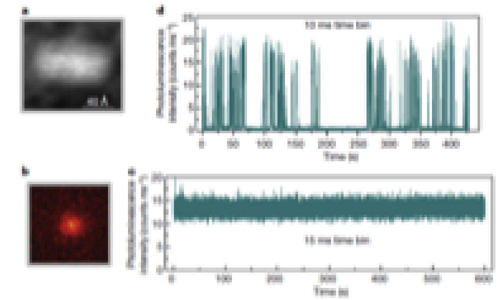
Key features are randomly distributed in time and space.

Key DLSR features:

High brilliance, quasi-CW: "unclocked" systems.

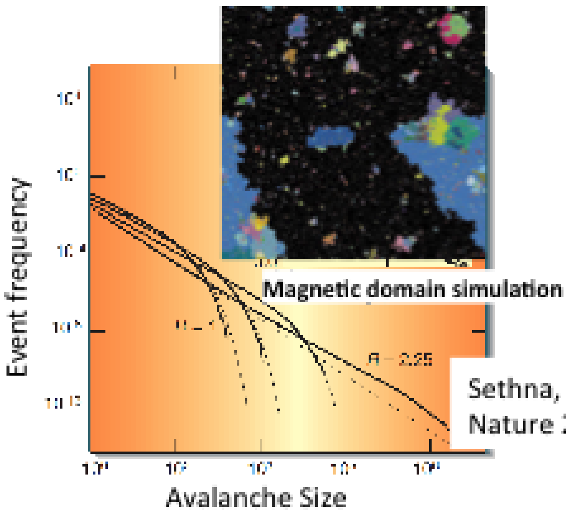
Simultaneously short length/time scales.

Optical Intermittency in Quantum Dots



Wang et al., Nature 2009

Domain-Wall Motion



Sethna, Nature 2001

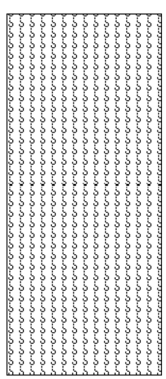
Unstructured Peptides and Amyloid Formation

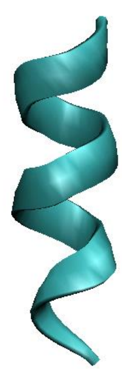
- Aggregation of unstructured peptides is linked to more than 15 neurodegenerative diseases – Alzeihmer's, Parkison's, Down's Syndrome, Huntington's etc, also Type II diabetes.
  - Affecting 4.5 million people in USA [1].
  - Health care cost > $50 billion[1].
- Industry – Protein aggregation during production, purification and storage.
  - Insulin production.

1 National Institute of Neurological Disorders and Stroke

GNNQQNY

Prion Disease





Amyloid β

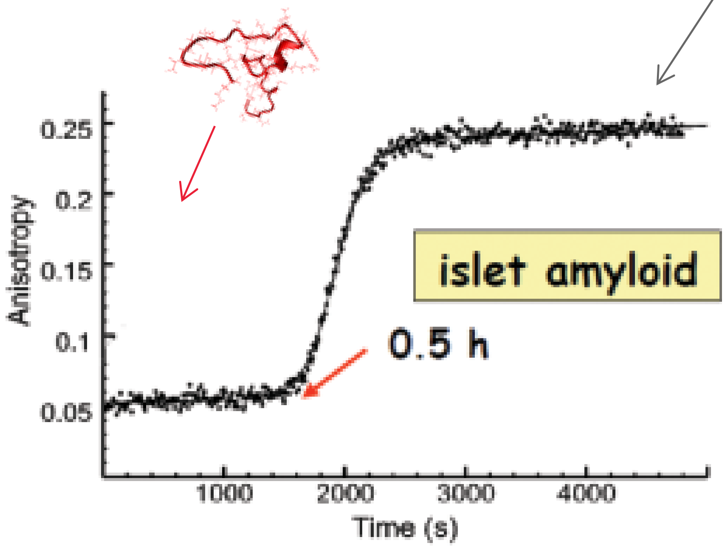
Alzheimer's Disease

Amylin (hIAPP)

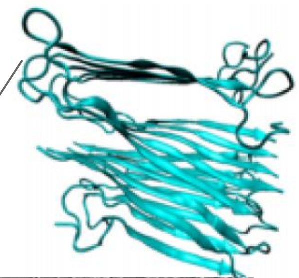
Type II Diabetes

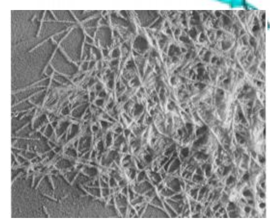
Juan de Pablo, University of Chicago

Nucleation and Growth of Toxic Aggregates



[Padrick SB et al. (2002) Biochem, 41:4694]

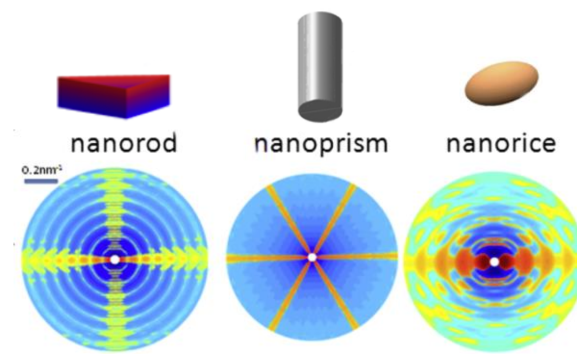




Shirahama and Cohen. JCB. 1967; 33 (3) 684

- Small aggregates
  - nucleate growth of fibrils
  - *extremely* toxic
- Structure unknown
- Nuclei formation time scales
  - μs to minutes
- The pathways for nuclei formation and growth are unknown.
- Monomer to oligomer to fibrils?

DLSR: Coherent scattering in heterogeneous systems during nucleation.

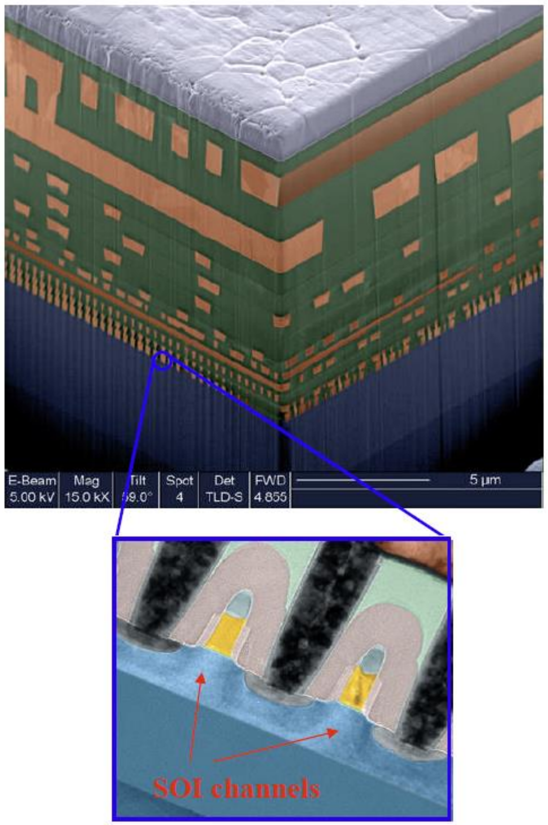


Simulation: shapes distinguished using coherent scattering

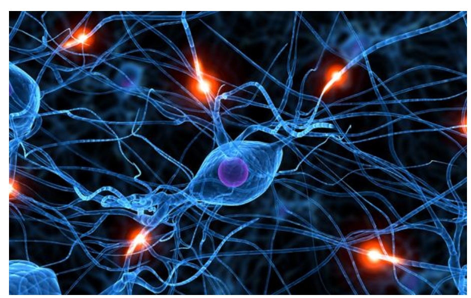
Chen, Zwart & Li, PRL 110 195501 (2013)

Juan de Pablo, University of Chicago

Integrating new functionalities in electronics



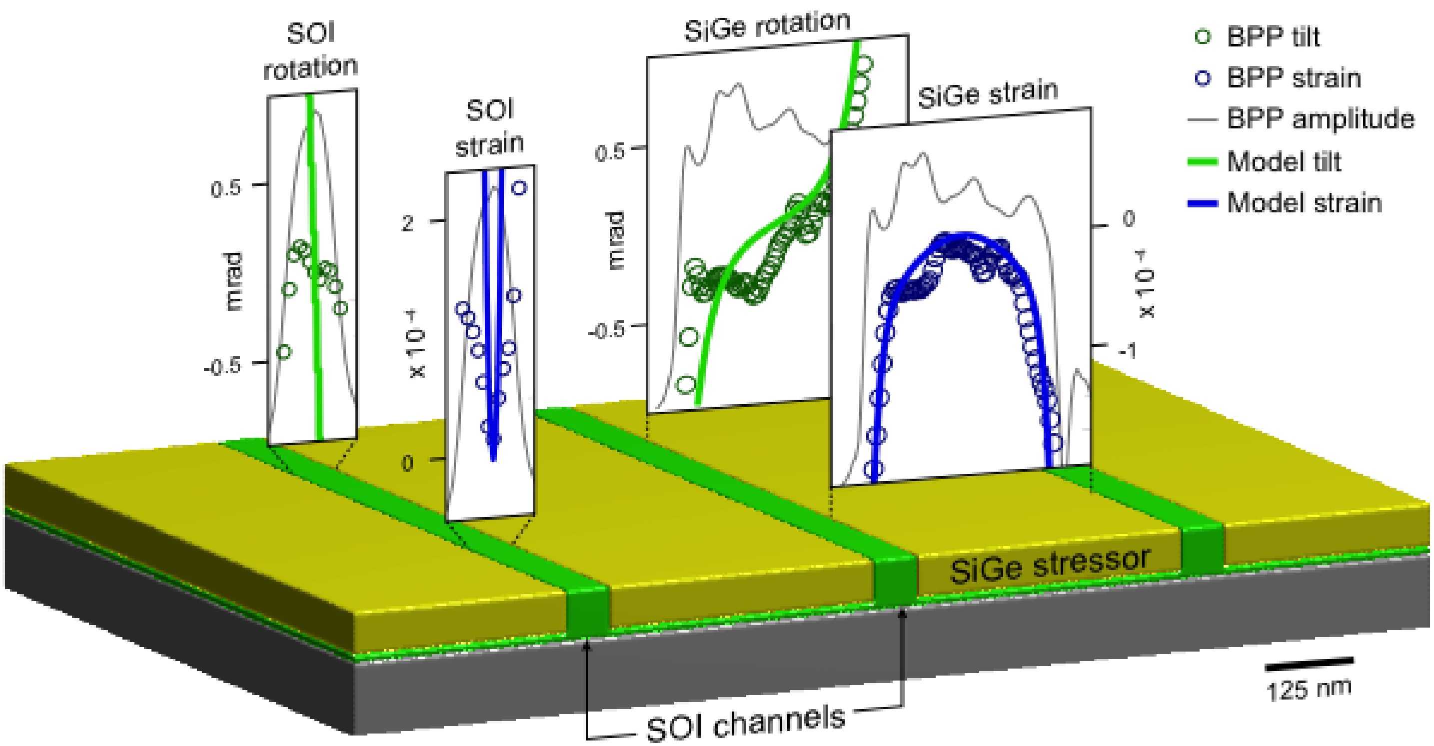
Today's electronics: Static structure of highly ordered crystalline devices.
Today's x-ray experiments match this. Multiple length scales problematic.



Advanced Electronics: New Functionalities

Future x-ray experiments: dynamics, heterogeneity, subtle order parameters.

Bragg ptychography of strain fields in devices



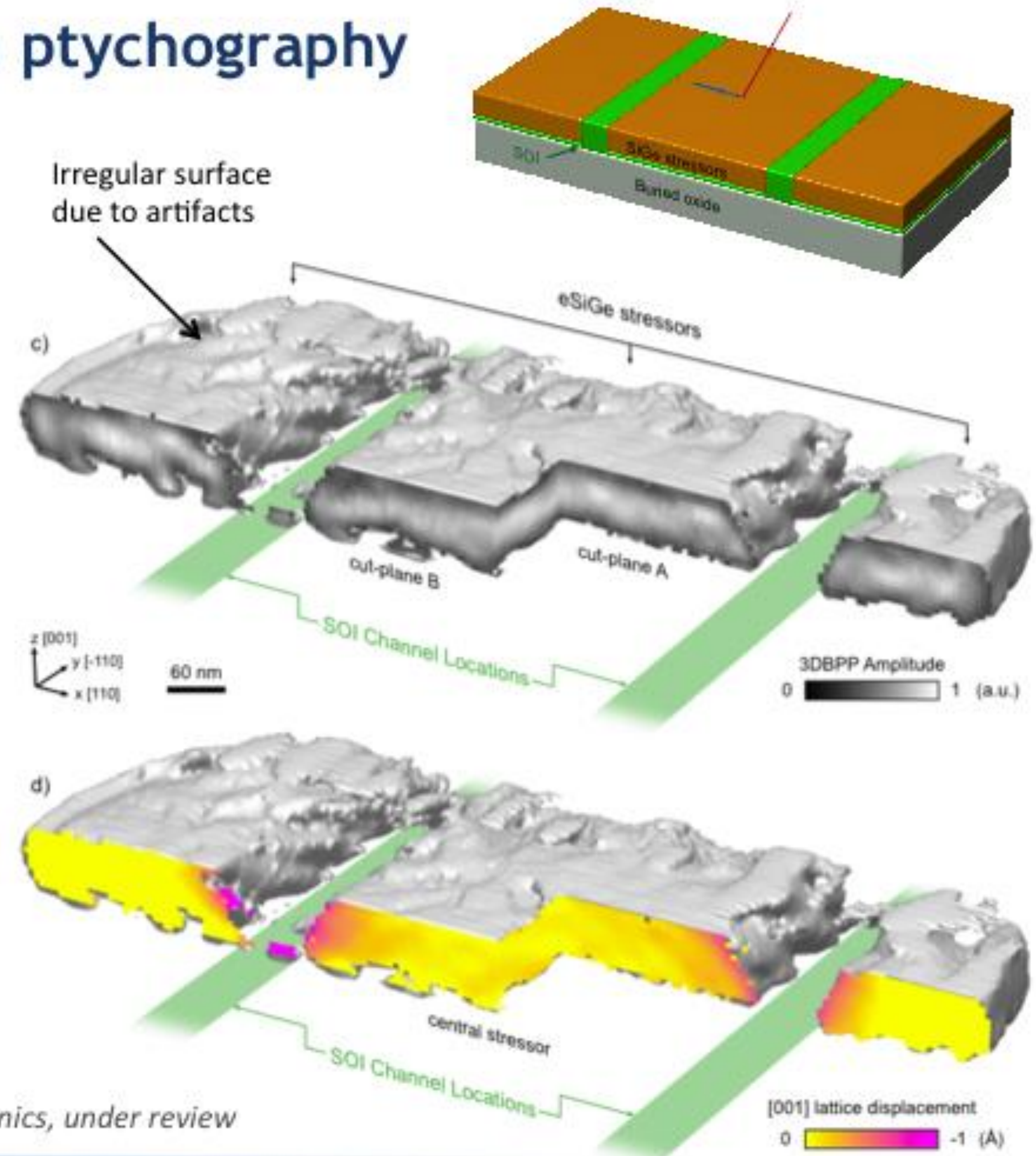
- Spatial resolution 6-20 nm
- Images of projected displacement field in 2D

BPP reconstructions consistent with linear elastic models of this stressor / SOI system.

Holt, et. al., PRL, 165502, 2014

3D Bragg projection ptychography

- 3D reconstructions are possible using 2D diffraction data.
- 3D imaging can be done at a lower dose and with a simpler experiment than previously possible.



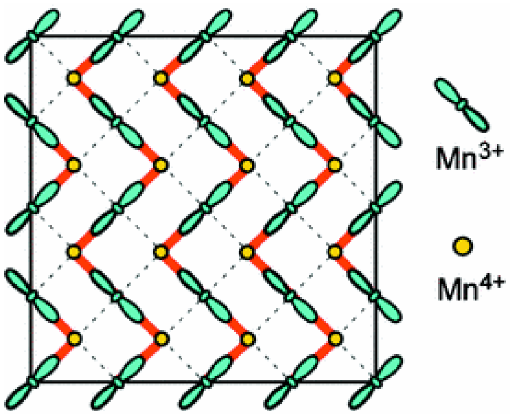
Internal lattice deformation at SiGe/SOI interfaces are revealed.

Hruszkewycz, et. al. , Nature Photonics, under review

Control of Other Degrees of Freedom

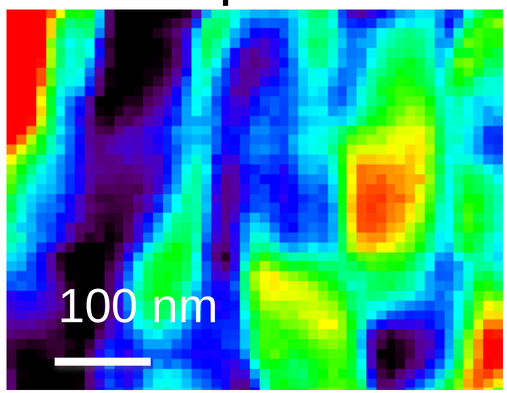
Future of information processing needs understanding and control of competing phases in the nano- to meso-scale regime

PCMO Electronic Orbital Phase



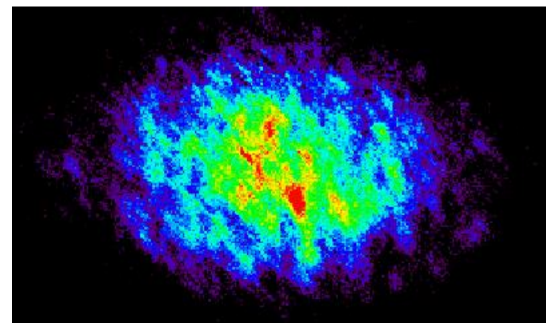
Today:

Real-Space Order



Resonant nanodiffraction from orbital order

Nanoscale Phase Fluctuations



Coherent x-ray speckle from nanoscale orbital order

- DLSRs enable enhancement in spatial resolution, 100x increase in sensitivity, access to ps timescales
- Will enable materials science and materials integration of new degrees of freedom.

Ian McNulty

Conclusion

- Technique Opportunities
  - New nanoprobes
  - Nanometer-scale microscopy with beam size matching problem
  - Improved time resolution in fluctuating systems
- Scientific implications span a vast range of problems.